

THEREFORE, WE CLAIM:

1. A crosslinking agent comprising an ungelled reaction product of the following reactants:

- 5 (A) at least one aminoplast resin;
- (B) a reactive urethane group-containing adduct which is a reaction product of the following reactants:
- (1) at least one polyisocyanate, and
- (2) at least one monohydric capping agent; and
- 10 (C) at least one compound different from (B) having active hydrogen groups reactive with aminoplast resin (A), said compound selected from at least one of:
- (i) compounds having the following structure (I):

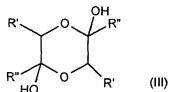
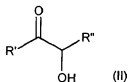


(I)

15 wherein X is aromatic; R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> can be the same or different and each independently represents H, (cyclo)alkyl having from 1 to 12 carbon atoms, aryl, alkaryl, aralkyl, or an active hydrogen-containing group,

20 provided that at least one of R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> represents an active hydrogen-containing group which is reactive with the aminoplast resin (A);

- (ii) compounds having the following structure (II) or (III):



where R' and R" are the same or different and each independently represents an aromatic group or an alkyl group having 1 to 12 carbon atoms; and

5 (iii) compounds different from (i) and (ii) and having a melting point of at least 80°C;

wherein said crosslinking agent is essentially free of urethane NH functionality and has a glass transition temperature of at least 10°C.

2. The crosslinking agent of claim 1, wherein the polyisocyanate (1) is  
10 selected from isocyanurate of isophorone diisocyanate trimer, isocyanurate of hexane diisocyanate trimer, and mixtures thereof.

3. The crosslinking agent of claim 2, wherein the polyisocyanate (1)  
15 comprises the isocyanurate of isophorone diisocyanate trimer.

4. The crosslinking agent of claim 1, wherein the monohydric capping  
agent (2) is selected from methanol, cyclohexanol, phenol, and mixtures  
thereof.

20 5. The crosslinking agent of claim 1, wherein the monohydric capping agent (2) comprises cyclohexanol.

6. The crosslinking agent of claim 1, wherein the ratio of NCO equivalents  
25 of the polyisocyanate (1) to OH equivalents of the monohydric capping agent (2) ranges from 0.8 to 1.0:1.

7. The crosslinking agent of claim 1, wherein the aminoplast resin (A) is  
or is derived from at least one of glycoluril, aminotriazine, and  
benzoguanamine.

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8. The crosslinking agent of claim 7, wherein the aminoplast resin (A) comprises alkoxyated aldehyde condensate of glycoluril.

9. The crosslinking agent of claim 8, wherein the aminoplast resin (A) comprises tetramethoxy methylglycoluril.

10. The crosslinking agent of claim 7, wherein the aminoplast resin (A) comprises (alkoxyalkyl) aminotriazine having one or less non-alkylated NH bond per triazine ring.

11. The crosslinking agent of claim 10, wherein the aminoplast resin (A) comprises (methoxymethyl) aminotriazine.

12. The crosslinking agent of claim 10, wherein the (alkoxyalkyl) aminotriazine has a degree of polymerization of 1.75 or less.

13. The crosslinking agent of claim 1, wherein the ratio of total combined moles of (B) and (C) to moles of aminoplast resin (A) ranges from 1 : 1.5 to 3.2

14. The crosslinking agent of claim 1, wherein the compound (C) comprises at least one compound having the structure (I).

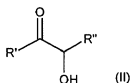
15. The crosslinking agent of claim 14, wherein at least one of R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> represents a group comprising an active hydrogen-containing group selected from hydroxyl, amide, amine, carboxylic acid, carbamate, urea, thiol, and mixtures thereof.

16. The crosslinking agent of claim 15, wherein at least one of R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> represents a group comprising at least one hydroxyl group.

17. The crosslinking agent of claim 16, wherein the compound (C) comprises benzyl alcohol.

5 18. The crosslinking agent of claim 1, wherein the compound (C) comprises at least one of compound (C)(ii).

19. The crosslinking agent of claim 18, wherein the compound (C) comprises a hydroxyl functional group-containing compound having the  
10 following structure (II):



or dimer derivatives thereof, wherein R' and R'' are the same or different and  
15 each independently represents an (cyclo)alkyl group having 1 to 12 carbon atoms or an aromatic group.

20 20. The crosslinking agent of claim 19, wherein one or both of R' and R'' represent aromatic groups.

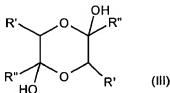
21. The crosslinking agent of claim 19, wherein the compound (C) comprises a compound selected from benzoin, hydroxycyclohexyl phenyl ketone, and mixtures thereof.

25 22. The crosslinking agent of claim 21, wherein the compound (C) comprises benzoin.

23. The crosslinking agent of claim 21, wherein the compound (C) comprises hydroxycyclohexyl phenyl ketone.

24. The crosslinking agent of claim 19, wherein one or both of R' and R'' are aromatic groups containing at least one heteroatom selected from furyl, pyridyl, methoxy phenyl, and dimethylaminophenyl groups.

25. The crosslinking agent of claim 18, wherein the active hydrogen group-containing compound (C) comprises a compound having the following structure (III):



wherein R and R' are the same or different and each independently represents an alkyl group having 1 to 12 carbon atoms or an aromatic group.

26. The crosslinking agent of claim 1, wherein the compound (C) comprises at least one of compound (C)(iii).

27. The crosslinking agent of claim 1, wherein compound (C) comprises an aliphatic mono-functional alcohol selected from borneol, norborneol, isoborneol, 1-adamantanemethanol, 1-adamantanol, 2-methyl-2-adamantanol and 5-norbornen-2-ol.

28. The crosslinking agent of claim 1, wherein the compound (C) is selected from benzoin, isoborneol, triphenylmethanol, N-tert-butylacrylamide, p-acetophenetidide, and mixtures thereof.

29. A method for preparing a crosslinking agent comprising the following steps:

(1) reacting the following reactants:

(a) at least one polyisocyanate, and

(b) at least one monohydric capping agent,

wherein the ratio of NCO equivalents of reactant (a) to OH equivalents of reactant (b) ranges from 0.8 to 1.0:1, to form a urethane group-containing reaction product;

(2) combining the following:

(A) at least one aminoplast resin;

(B) the urethane group-containing reaction product formed in step (1); and

(C) at least one compound different from (B) having active hydrogen groups reactive with aminoplast resin (A), said compound selected from at least one of:

(i) compounds having the following structure (I):

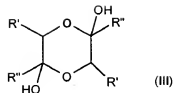
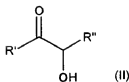


(I)

wherein X is aromatic; R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> can be the same or different and each independently represents H, (cyclo)alkyl having from 1 to 12 carbon atoms, aryl, alkaryl, aralkyl, or an active hydrogen-containing group,

provided that at least one of R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> represents an active hydrogen-containing group which is reactive with the aminoplast resin (A);

(ii) compounds having the following structure (II) or (III):



where R' and R'' are the same or different and each independently represents an aromatic group or an alkyl group having 1 to 12 carbon atoms; and

- 5 (iii) compounds different from (i) and (ii) and having a melting point of at least 80°C;

in a ratio of total combined moles of (B) and (C) to moles of aminoplast resin (A) ranging from 1 : 1.5 to 3.2 to form a reaction admixture; and

- (3) heating the reaction admixture formed in step (2) to a  
10 temperature ranging from 95°C to 135°C for a time sufficient to form a crosslinking agent having a glass transition temperature of at least 25°C which is essentially free of urethane NH functionality as determined by infrared spectroscopy.

- 15 30. The method of claim 29, wherein the polyisocyanate (a) is selected from isocyanurate of isophorone diisocyanate trimer, isocyanurate of hexane diisocyanate trimer, and mixtures thereof.

31. The method of claim 30, wherein the polyisocyanate (a) comprises the  
20 isocyanurate of isophorone diisocyanate trimer.

32. The method of claim 29, wherein the monohydric capping agent (b) is selected from methanol, cyclohexanol, phenol, and mixtures thereof.

- 25 33. The method of claim 32, wherein the monohydric capping agent (b) comprises cyclohexanol.

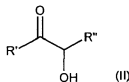
34. The method of claim 29, wherein the aminoplast resin (A) is or is derived from at least one of glycoluril, aminotriazine, and benzoguanamine.
35. The method of claim 34, wherein the aminoplast resin (A) comprises alkoxyalated aldehyde condensate of glycoluril.
36. The method of claim 35, wherein the aminoplast resin (A) comprises tetramethoxy methylglycoluril.
37. The method of claim 34, wherein the aminoplast resin (A) comprises (alkoxyalkyl) aminotriazine having one or less non-alkylated NH bond per triazine ring.
38. The method of claim 37, wherein the aminoplast resin (A) comprises (methoxymethyl) aminotriazine.
39. The method of claim 37, wherein the (alkoxyalkyl) aminotriazine has a degree of polymerization of 1.75 or less.
40. The method of claim 29, wherein the compound (C) comprises at least one compound having the structure (I).
41. The method of claim 40, wherein at least one of R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> represents a group comprising an active hydrogen-containing group selected from hydroxyl, amide, amine, carboxylic acid, carbamate, urea, thiol, and mixtures thereof.
42. The method of claim 41, wherein at least one of R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> represents a group comprising at least one hydroxyl group.



43. The method of claim 29, wherein compound (C) comprises benzyl alcohol.

44. The method of claim 29, wherein the compound (C) comprises at least one of compound (C)(ii).

45. The method of claim 44, wherein the compound (C) comprises a hydroxyl functional group-containing compound having the following structure (II):



or dimer derivatives thereof, wherein R' and R'' are the same or different and each independently represents a (cyclo)alkyl group having 1 to 12 carbon atoms or an aromatic group.

46. The method of claim 45, wherein one or both of R' and R'' represent aromatic groups.

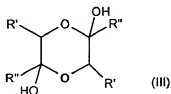
47. The method of claim 45 wherein compound (C) comprises a compound selected from benzoin, hydroxycyclohexyl phenyl ketone, and mixtures thereof.

48. The method of claim 47, wherein the compound (C) comprises benzoin.

49. The method of claim 47, wherein the compound (C) comprises hydroxycyclohexyl phenyl ketone.

50. The method of claim 45, wherein one or both of R' and R'' are aromatic groups containing at least one heteroatom selected from furyl, pyridyl, methoxy phenyl, and dimethylaminophenyl groups.

- 5 51. The method of claim 44, wherein compound (C) comprises a compound having the following structure (III):



- 10        wherein R' and R'' are the same or different and each independently represents an alkyl group having 1 to 12 carbon atoms or an aromatic group.

- 12 52. The method of claim 29, wherein the compound (C) comprises at least one of compound (C)(iii).

- 15 53. The method of claim 52, wherein compound (C) comprises an aliphatic mono-functional alcohol selected from borneol, norborneol, isoborneol, 1-adamantanemethanol, 1-adamantanol, 2-methyl-2-adamantanol, and 5-norbornen-2-ol.

- 20 54. The method of claim 29, wherein the compound (C) comprises a compound selected from benzoin, isoborneol, triphenylmethanol, N-tert-butylacrylamide, p-acetophenetidine, and mixtures thereof.

- 25 55. A curable composition comprising the following components:  
(A) a film-forming polymer having reactive functional groups; and

(B) a crosslinking agent having functional groups reactive with the functional groups of (A),

said crosslinking agent comprising an ungelled reaction product of the following:

(1) at least one aminoplast resin;  
(2) a reactive urethane group-containing adduct which is a reaction product of the following reactants:

(a) at least one polyisocyanate, and  
(b) at least one monohydric capping agent;  
(3) at least one compound different from (2) having active hydrogen groups reactive with aminoplast resin (1), said compound selected from at least one of:

(i) compounds having the following structure (I):

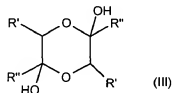
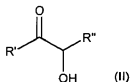


(I)

wherein X is aromatic; R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> can be the same or different and each independently represents H, (cyclo)alkyl having from 1 to 12 carbon atoms, aryl, alkaryl, aralkyl, or an active hydrogen-containing group,

provided that at least one of R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> represents an active hydrogen-containing group which is reactive with the aminoplast resin (A);

(ii) compounds having the following structure (II) or (III):



where R' and R'' are the same or different and each independently represents an aromatic group or an alkyl group having 1 to 12 carbon atoms; and

(iii) compounds different from (i) and (ii) and having a melting point of at least 80°C;

wherein said crosslinking agent is essentially free of urethane NH functionality.

56. The curable composition of claim 55, wherein the polymer (A) is selected from acrylic, polyester, polyepoxide polyurethane and polyether polymers, and mixtures thereof.

57. The curable composition of claim 55, wherein the polymer (A) comprises reactive functional groups selected from the group consisting of hydroxyl, carbamate, epoxy, carboxylic acid, and mixtures thereof.

58. The curable composition of claim 57, wherein the polymer (A) comprises hydroxyl and/or carbamate functional groups.

59. The curable composition of claim 57, wherein the polymer (A) comprises hydroxyl and/or epoxy functional groups.

60. The curable composition of claim 55, wherein the polymer (A) is present in an amount ranging from 90 to 5 weight percent based on weight of total resin solids present in the composition.

61. The curable composition of claim 55, wherein the polyisocyanate (a) is selected from isocyanurate of isophorone diisocyanate trimer, isocyanurate of hexane diisocyanate trimer, and mixtures thereof.

62. The curable composition of claim 61, wherein the polyisocyanate (a) comprises an isocyanurate of isophorone diisocyanate trimer.

5 63. The curable composition of claim 55, wherein the monohydric capping agent (b) is selected from methanol, cyclohexanol, phenol, and mixtures thereof.

64. The curable composition of claim 63, wherein the monohydric capping agent (b) comprises cyclohexanol.

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65. The curable composition of claim 55, wherein the ratio of NCO equivalents of the polyisocyanate (a) to OH equivalents of the monohydric capping agent (b) ranges from 0.8 to 1.0:1.

15 66. The curable coating composition of claim 55, wherein the aminoplast resin (A) is or is derived from at least one of glycoluril, aminotriazine, and benzoguanamine.

20 67. The curable coating composition of claim 66, wherein the aminoplast resin (A) comprises an alkoxyated aldehyde condensate of glycoluril.

68. The curable powder composition of claim 67, wherein the aminoplast resin (A) comprises tetramethoxy methylglycoluril.

25 69. The curable powder coating composition of claim 66, wherein the aminoplast resin (A) comprises (alkoxyalkyl) aminotriazine having one or less non-alkylated NH bond per triazine ring.

30 70. The curable powder coating composition of claim 69, wherein the aminoplast resin (A) comprises (methoxymethyl) aminotriazine.

71. The curable powder coating composition of claim 69, wherein the (alkoxyalkyl) aminotriazine has a degree of polymerization of 1.75 or less.

5 72. The curable composition of claim 55, wherein the ratio of total combined moles of (2) and (3) to moles of aminoplast resin (1) ranges from 1:1.5 to 3.2.

73. The curable composition of claim 55, wherein the compound (C) comprises at least one compound having the structure (I).

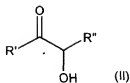
74. The curable composition of claim 73, wherein at least one of  $R^1$ ,  $R^2$ , and  $R^3$  represents a group comprising an active hydrogen-containing group selected from hydroxyl, amide, amine, carboxylic acid, carbamate, urea, thiol, and mixtures thereof.

75. The curable composition of claim 73, wherein at least one of  $R^1$ ,  $R^2$ , and  $R^3$  represents a group comprising at least one hydroxyl group.

76. The curable composition of claim 73, wherein compound (C) comprises benzyl alcohol.

77. The curable composition of claim 55, wherein the compound (C) comprises at least one of compound (C)(i).

78. The curable composition of claim 77, wherein the compound (C) comprises a hydroxyl functional group-containing compound having the following structure (II):



or dimer derivatives thereof, wherein R' and R'' are the same or different and each independently represents an (cyclo)alkyl group having 1 to 12 carbon atoms or an aromatic group.

79. The curable composition of claim 78, wherein one or both of R' and R'' represent aromatic groups.

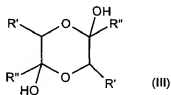
80. The curable composition of claim 78, wherein compound (C) comprises a compound selected from benzoin, hydroxycyclohexyl phenyl ketone, and mixtures thereof.

81. The curable composition of claim 80, wherein the compound (C) comprises benzoin.

82. The curable composition of claim 80, wherein the compound (C) comprises hydroxycyclohexyl phenyl ketone.

83. The curable composition of claim 78, wherein one or both of R and R' are aromatic groups containing at least one heteroatom selected from furyl, pyridyl, methoxy phenyl, and dimethylaminophenyl groups.

84. The curable composition of claim 77, wherein compound (C) comprises a compound having the following structure (III):



wherein R' and R'' are the same or different and each independently represents an alkyl group having 1 to 12 carbon atoms or an aromatic group.

85. The curable composition of claim 55, wherein the compound (C) comprises a compound selected from aliphatic monofunctional alcohols, aryl aliphatic mono-functional alcohols, secondary amides, aliphatic open-chain amides, aromatic (poly)cyclic amides, aromatic open-chain amide, and monofunctional urethanes.

86. The curable composition of claim 85, wherein compound (C) comprises an aliphatic mono-functional alcohol selected from borneol, norborneol, isoborneol, 1-adamantanemethanol, 1-adamantanol, 2-methyl-2-adamantanol, and 5-norbornen-2-ol.

87. The curable composition of claim 55, wherein the compound (C) comprises a compound selected from benzoin, isoborneol, triphenylmethanol, N-tert-butylacrylamide, p-acetophenetidine, and mixtures thereof.

88. The curable composition of claim 55, wherein the crosslinking agent (B) is present in an amount ranging from 5 to 95 weight percent based on weight of total resin solids present in the composition.

89. The curable composition of claim 55, wherein said composition is a curable powder coating composition.

90. A multilayer composite coating composition comprising a pigmented base coat deposited from a base coat film-forming composition and a transparent top coat over at least a portion of the base coat, said top coat deposited from a curable powder top coating composition comprising a solid particulate film-forming mixture of the following components:



(A) a polymer containing reactive functional groups, said polymer having a glass transition temperature of at least 30°C; and

(B) a crosslinking agent having functional groups reactive with the functional groups of (A), said crosslinking agent comprising an ungelled reaction product of the following:

(1) at least one aminoplast resin;

(2) a reactive urethane group-containing adduct which is a reaction product of the following reactants:

(a) at least one polyisocyanate, and

(b) at least one monohydric capping agent; and

(3) at least one compound different from (2) having active hydrogen groups reactive with aminoplast resin (1), said compound selected from at least one of:

(i) compounds having the following structure (I):



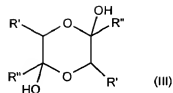
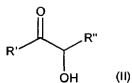
(I)

wherein X is aromatic; R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> can be the same or different and each independently represents H, (cyclo)alkyl having from 1 to 12 carbon atoms, aryl, alkaryl, aralkyl, or an active hydrogen-containing group,

provided that at least one of R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> represents an active hydrogen-containing group which is reactive with the aminoplast resin (A);

(ii) compounds having the following structure (II) or

(III):



where R' and R'' are the same or different and each independently represents an aromatic group or an alkyl group having 1 to 12 carbon atoms; and

- 5 (iii) compounds different from (i) and (ii) and having a melting point of at least 80°C;

wherein said crosslinking agent is essentially free of urethane NH functionality and has a glass transition temperature of at least 10°C.

- 10 91. A substrate coated with the powder coating composition of claim 89.
92. A substrate coated with the curable composition of claim 55.
- 15 93. A substrate coated with the multilayer coating composition of claim 90.